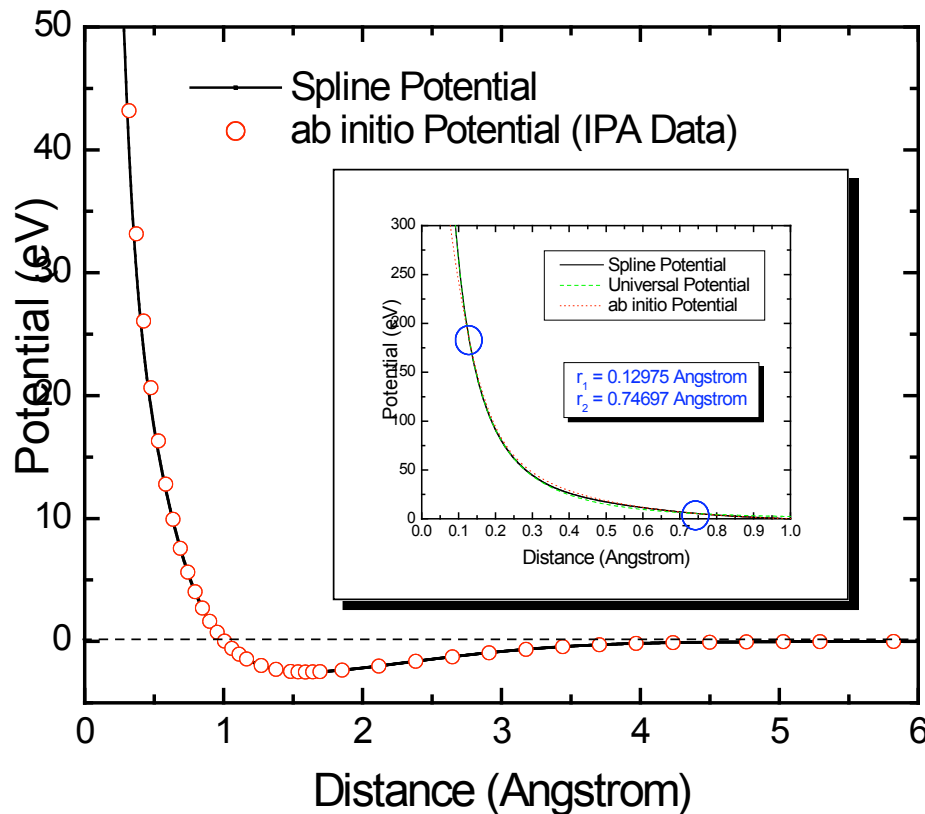


Molecular Dynamics Calculations on Hydrogenated and Pure Liquid Lithium Surfaces

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H-Li Inter-Atomic Potential



- H-Li potential combines:
 - singlet *ab initio* potential [1,2]
 - universal potential at small distance
- Spline b/w 0.37 Å ~ 0.7565 Å
- Interaction Cutoff: 5 Å
- Bond Min Distance: 1.05 Å
- Bond Max Distance: 3.60 Å
- Bond Energy: 2.526 eV
- Weight function [3] :

$$f_c(r) = \frac{1}{2} + \frac{1}{2} \cos\left[\frac{\pi(r - r_1)}{r_2 - r_1}\right]$$

[1] N. Geum, et al., *Interaction potentials of LiH, NaH, KH, RbH and CsH*, J. Chem. Phys., **115** (2001) 5984.

[2] R. Cote, et al., *Enhanced Cooling of Hydrogen Atoms by Lithium Atoms*, Physical Review Letters, **84** (2000) 2806.

[3] R. Taylor, et al., *Molecular Dynamics Simulations of Reactions between Molecules*, Langmuir, **11** (1995) 1220.

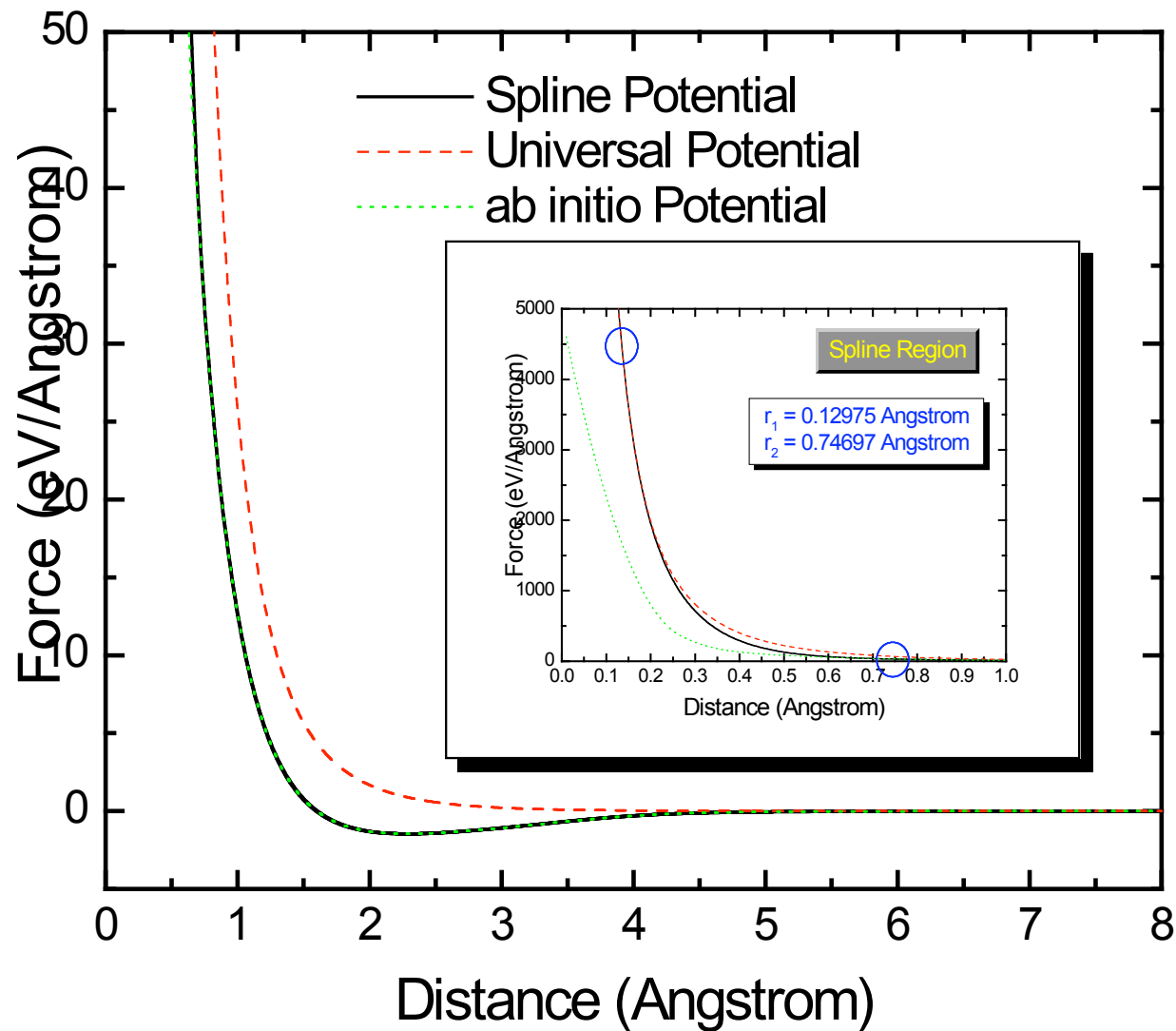
Why Trust It?

- Based on the Inverted Perturbation Approach (IPA) potential ^[1]
- Includes dispersion forces and exchange interactions
- Uses a large scale configuration interaction method
- Calculated by MOLCAS program ^[4], a specific program used for molecular configuration-interaction calculations
- Enable accurate predictions to be made of the scattering of the alkali metal atoms by hydrogen atoms ^[1,2]
- The resulting interaction potential is available to public from AIP ^[5]

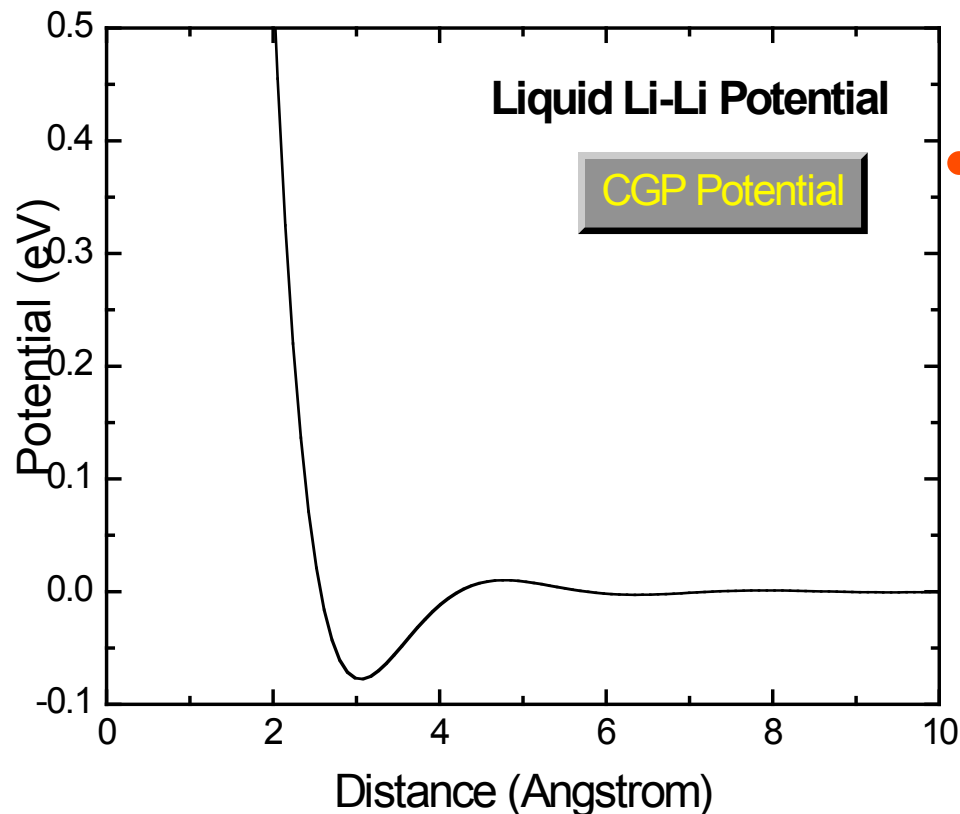
^[4] K. Anderson, M. R. A. Blomberg, M. P. Fulscher *et al.*, *MOLCAS*, Version 4, 1997, Lund University, Sweden.

^[5] American Institute of Physics (AIP), EPAPS <http://www.aip.org/pubservs/epaps.html>

H-Li Inter-Atomic Force



Liquid Li-Li CGP Potential



• The fluctuations around 5 Å show the long-range interaction

- Interaction Cutoff: 5.75 Å
- Bond Min Distance: 2.60 Å
- Bond Max Distance: 4.13 Å
- Bond Energy: 0.083 eV

[6] Manel Canales, L. E. Gonzalez, J. A. Padro, *Computer Simulation Study of Liquid Lithium at 470 and 843K*, *Physical Review E*, **50** (1994) 3656.

Why Trust This Li-Li Potential?

- Obtained from the Neutral Pseudoatom (NPA) method [6]
- Calculated by a specific MD group for liquid Li of Spain [6]
- Involving both structural and thermodynamic properties for liquid Lithium
- Reasonable agreement with available experimental data [6]
- Effective for high temperature ranging 470K ~ 843K

MolDyn Code

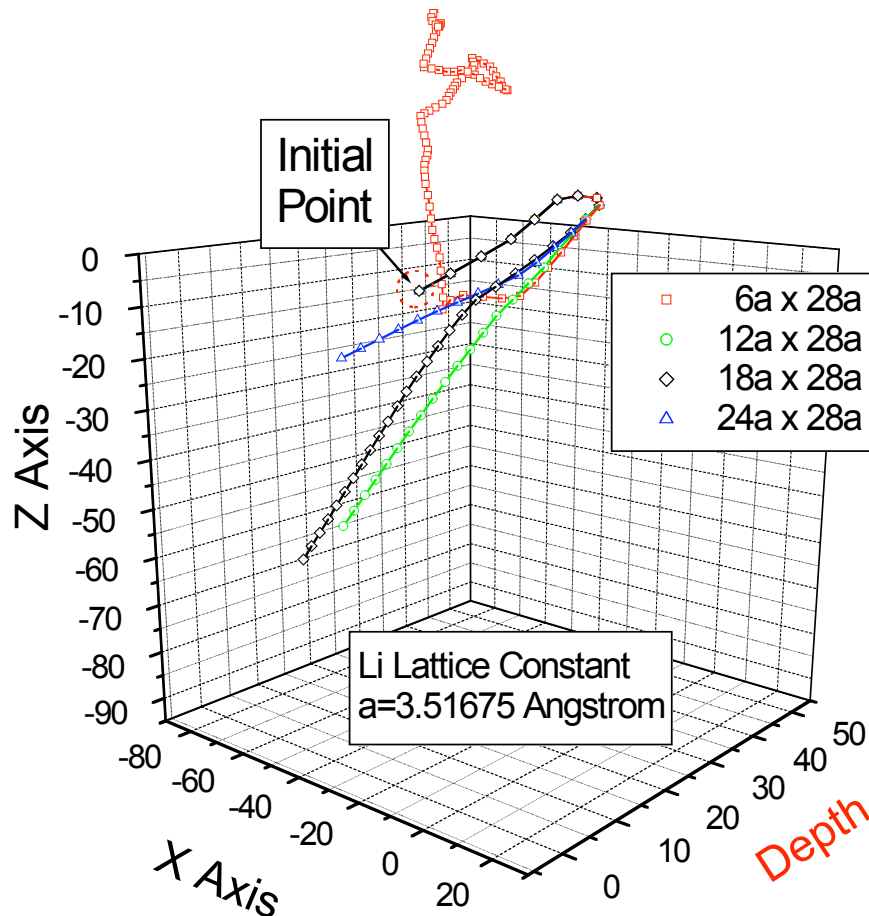
- Molecular Dynamics code (MolDyn)
 - Fixed timestep Beeman integrator ^[7]
 - Berendsen method to control temperature ^[8]
 - Modified to handle lithium/hydrogen systems
- Code configurations
 - **Time step:** 0.1 fs
 - **Dimensions:** 42Å x 42Å x 60Å (deep)
 - Lattice size: 5040 Li atoms
 - Time scale: run for 2000 fs
 - Periodical side boundary and fixed bottom
 - Cutoff: 5.75 Å for Li-Li & 5 Å for D-Li
- Creation of the pure hydrogenated liquid Li lattice
 - Originally from BCC structure
 - Slowly heated to required temperature
- Creation of the liquid Li lattice
 - Initially build a pure lithium lattice at a concerned temperature
 - Allows the development of a more realistic hydrogenated surface with 1000s of individual H/D impacts
 - The generated H:Li surface is used as the input in all next subsequent runs to calculate concerned info

^[7] D. Beeman, *J. Comp. Phys.*, **20** (1976) 130.

^[8] H. J. C. Berendsen, *et al.*, *J. Chem. Phys.*, **81** (1984) 3684.

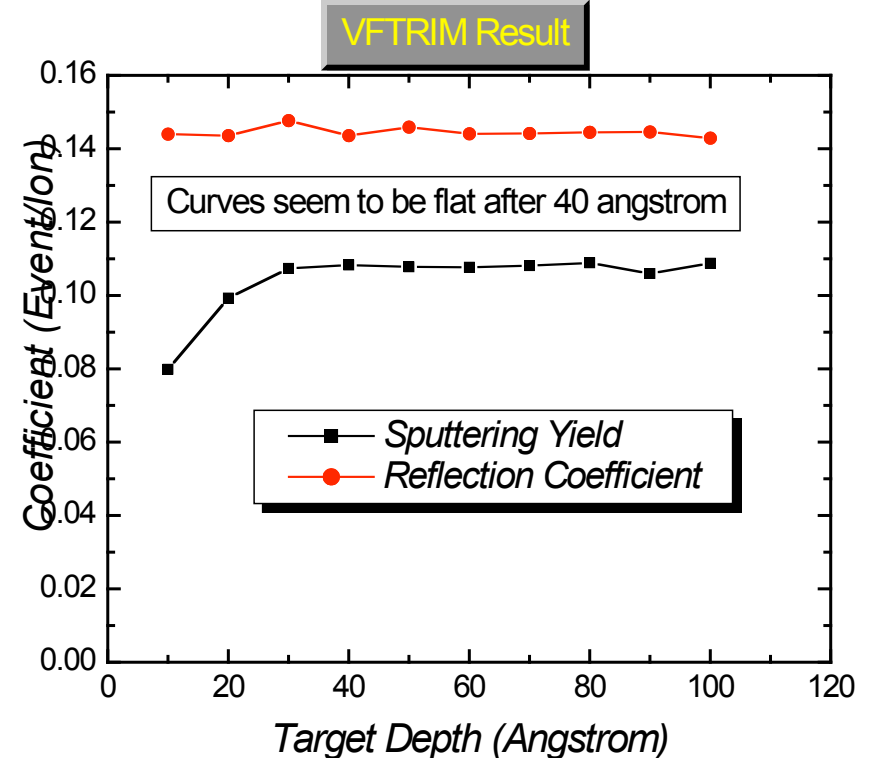
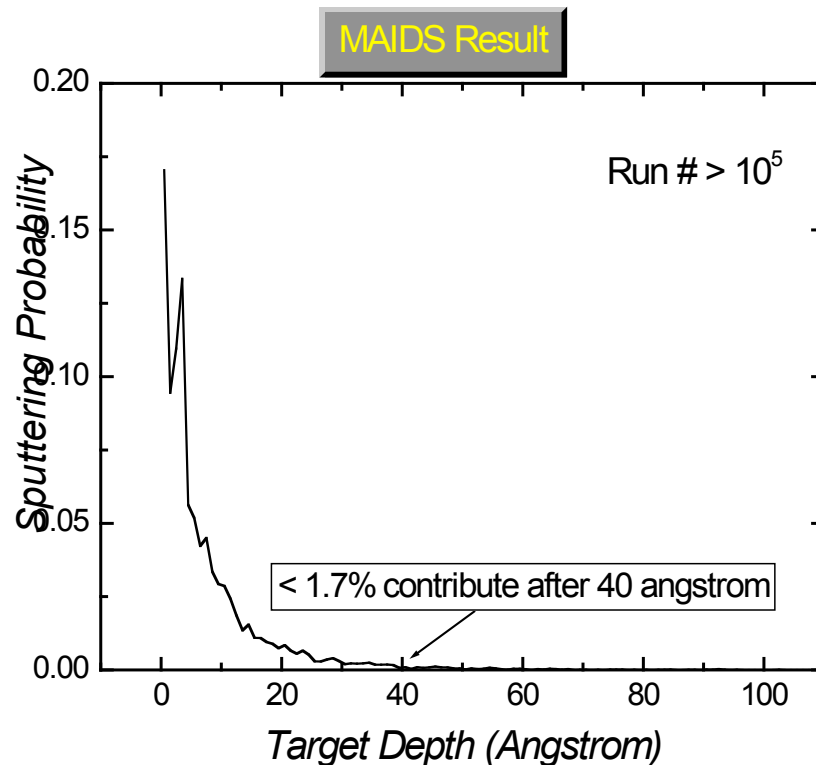
Preparation of Liquid Li Surface

-- Surface Dimensions XZ



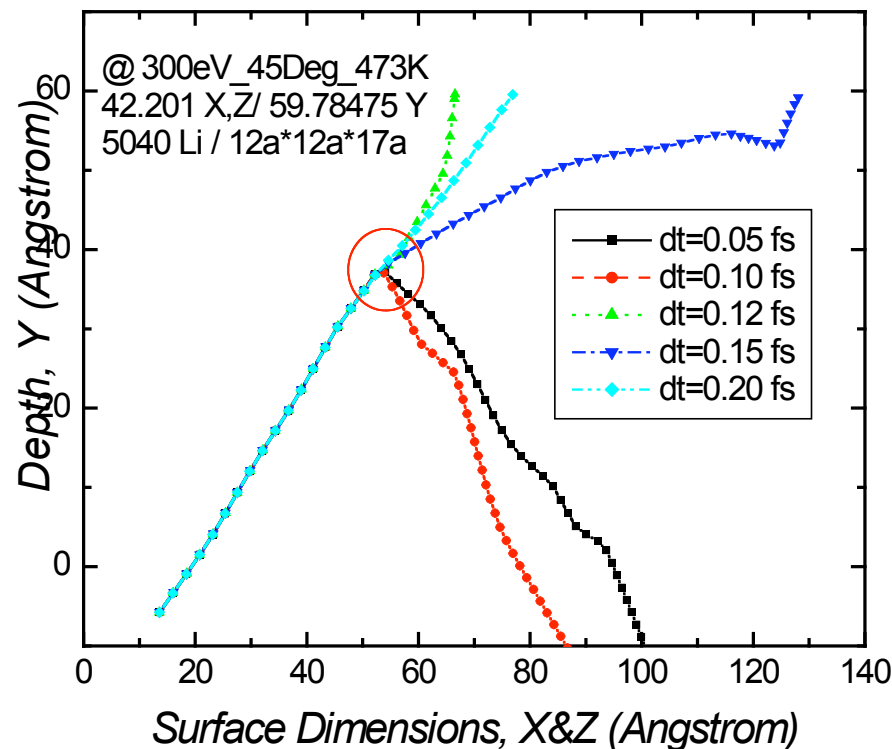
- Surface dimension (x,z) must be greater than twice cutoff (11.5\AA)
 - Insures that an atom does not interact with an image of itself, and interacts with only one image of every other atom
- We choose 42\AA , about eight times of cutoff distance as lattice surface dimension
- Increasing x,z dimensions does not affect incident ion trajectories
- Test for this XZ dimensions
 - H_Li_300eV_045°_045°_0K_dt0.1fs
 - Got to be reflected in the end
 - $XZ = 12a = 42.201\text{\AA}$ seems to be good enough

Preparation of Liquid Li Surface -- Depth Dimension



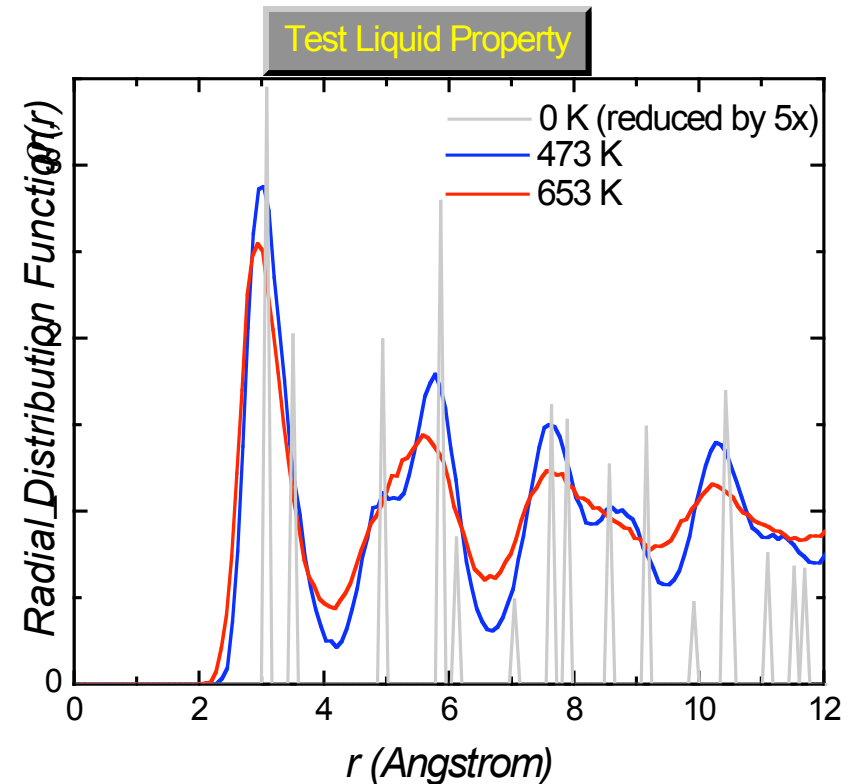
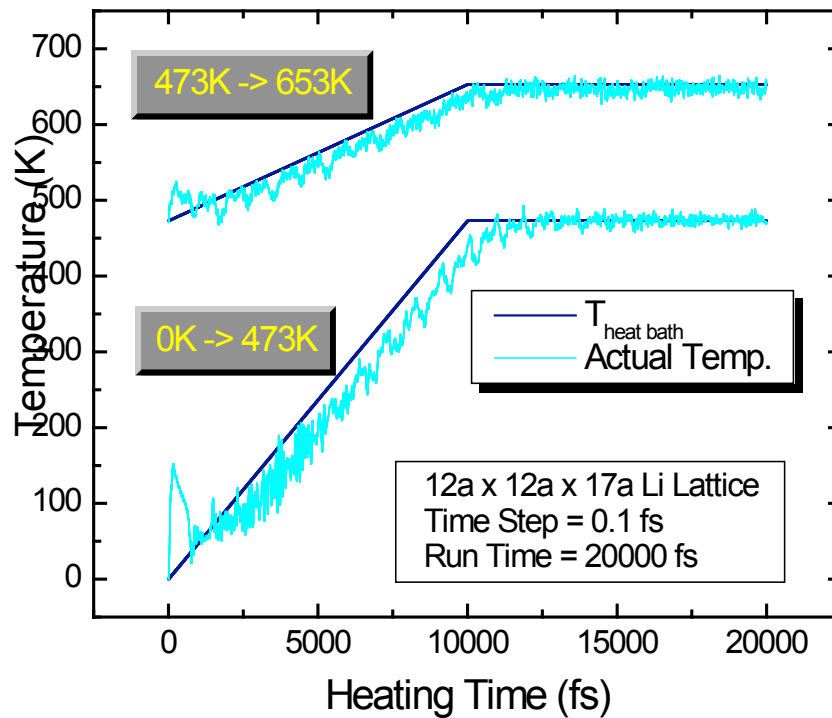
- MAIDS implies the deepest sputtered atom comes from 40Å
- VFTRIM shows no effect on sputtering yield by making the depth larger than ~40 Å
- Taking account for cascade effect, we choose 60Å as lattice depth

Preparation of Liquid Li Surface -- Time Step



- $dt > 0.1$ fs gives an incorrect trajectory for a 300 eV H collision with a Li surface atom
- $dt \leq 0.1$ fs seems good enough to model the backscatter & sputtering
- As a tradeoff (accuracy vs. CPU time), **dt = 0.1 fs** is a good choice

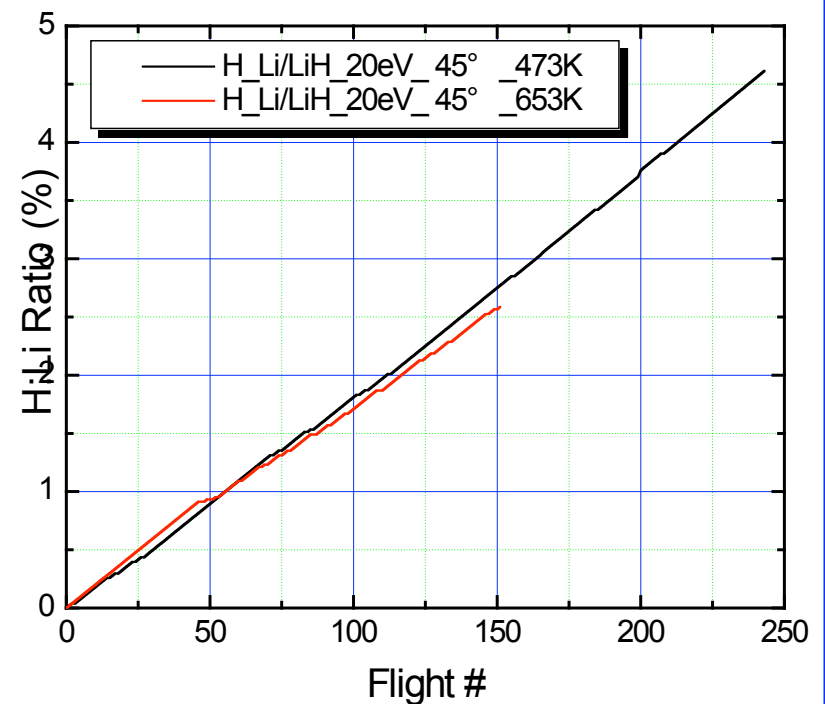
Creation of Pure Liquid Li Surface



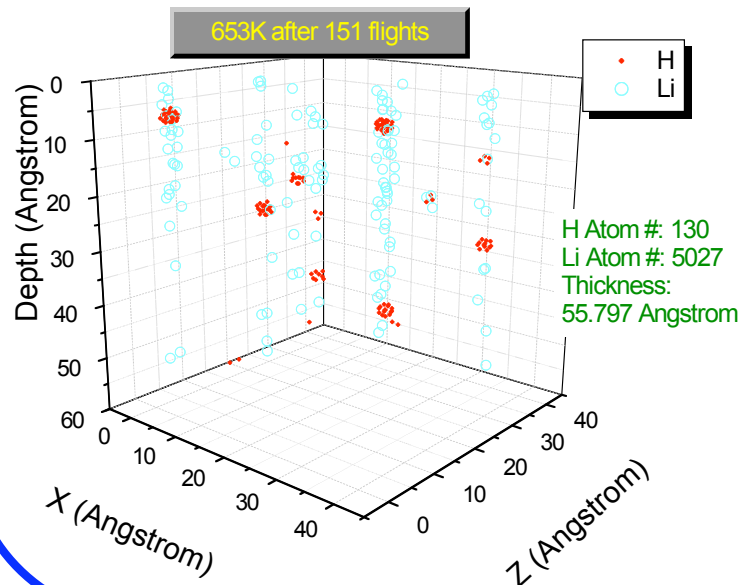
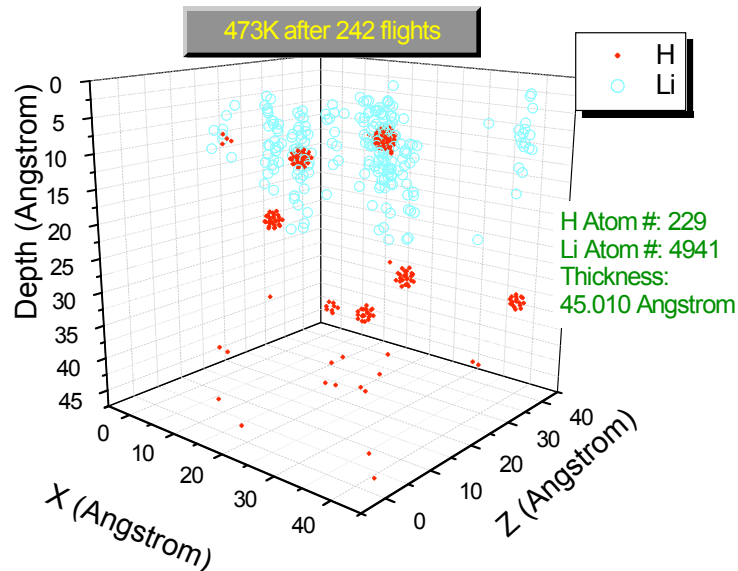
- Heat is added gradually over first 10,000 fs
- The surface is allowed to equilibrate for another 10,000 fs
- The radial distribution function of solid (BCC) structure is discrete
- The oscillating radial distribution curves prove liquid property of the created surfaces

Hydrogenated Liquid Li Surface

- Continuously incident H on the hydrogenated liquid Li surfaces
- Detailed cases
 - H_Li/LiH_20eV_45°_473K
 - H_Li/LiH_20eV_45°_653K
- Results
 - Sputtering and Reflection
 - Hydride concentration H:Li
 - Implantation characterization of H



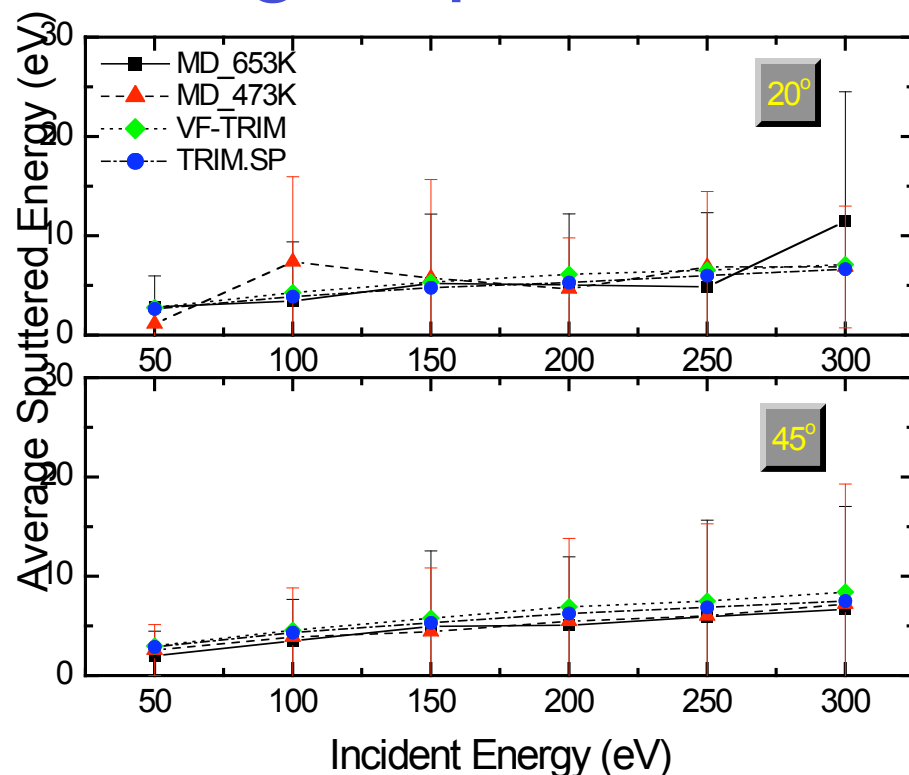
Lithium Hydride Concentration



- H:Li ratio in initial stage linearly increases before saturation
- Saturation is not obtained due to the insufficient time scale (2 ps)
- The actual concentration can not stay at such a high level according to the phase diagram with no H pressure on surface
- H atoms preferably diffuse towards the other H atoms within around layers in similar depth, H-H bond energy > Li-H bond energy.
- H atoms are more likely to bond together with H atoms
- Thickness decreases due to removal of sputtered Li atoms

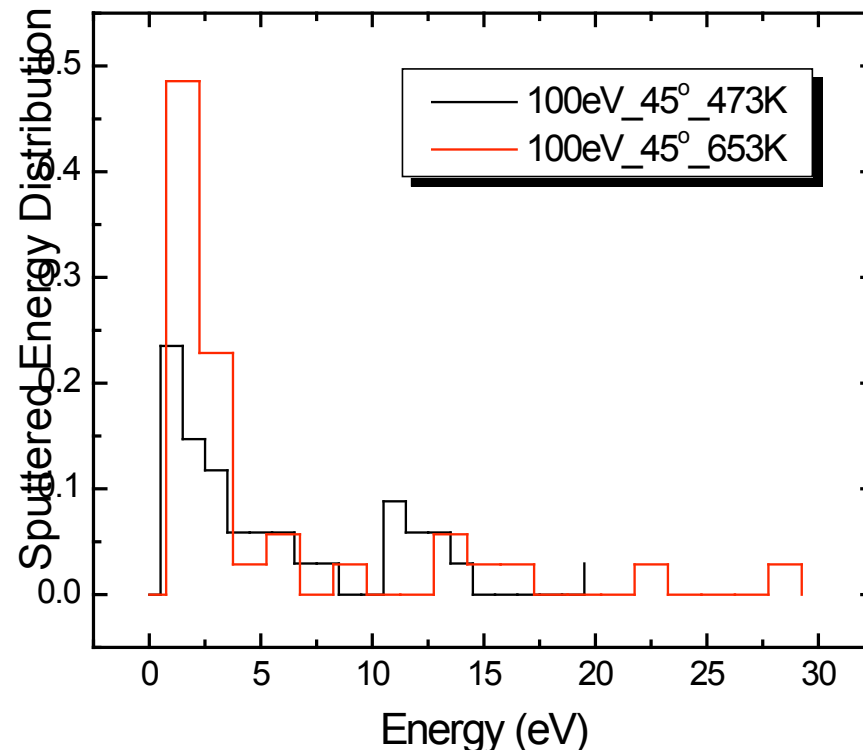
Sputtering Results on Pure Li Surface

-- Average Sputtered Energy



- Surface temperature has little effect on energy of sputtered atoms in MD
- Incident angle seems not to effect the energy of the sputtered atoms
- MD and VF-TRIM & TRIM.SP show better agreement at low energies
- The average energy of the sputtered Li atoms is in the order of several eV's

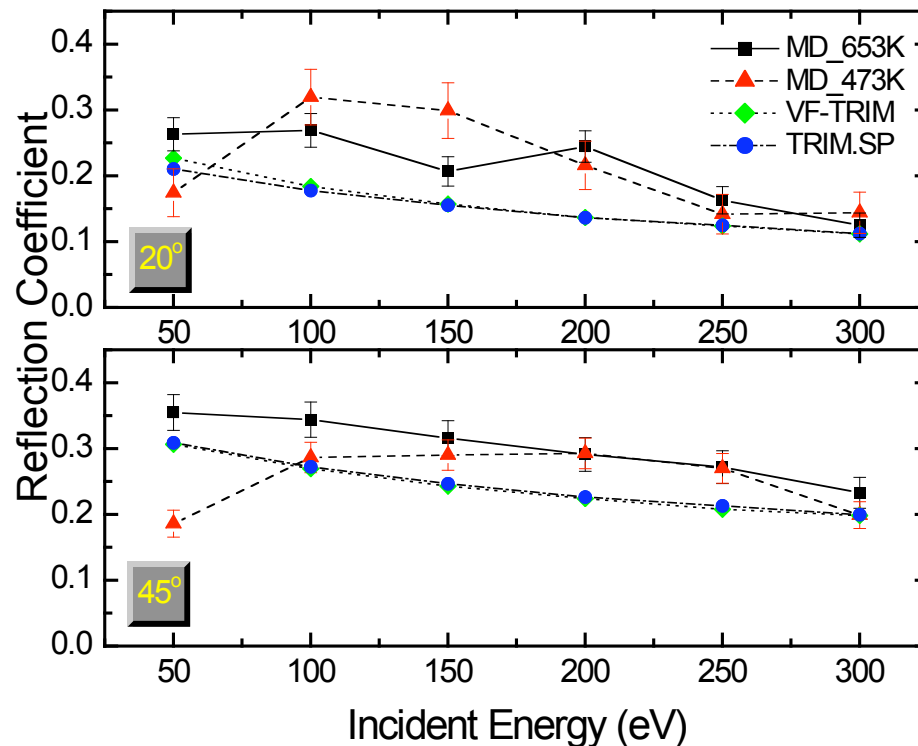
Sputtered Energy Distribution & Vapor Pressure



- There are two peaks for the energy distribution of sputtered atoms
- Higher surface temperature leads to significantly increasing of vaporization flux
- Low-energy peak corresponds to simulated-evaporation component
- Calculate vapor pressure of the liquid Li surface to see if it can be seen in MD

Reflection Results on Pure Li Surface

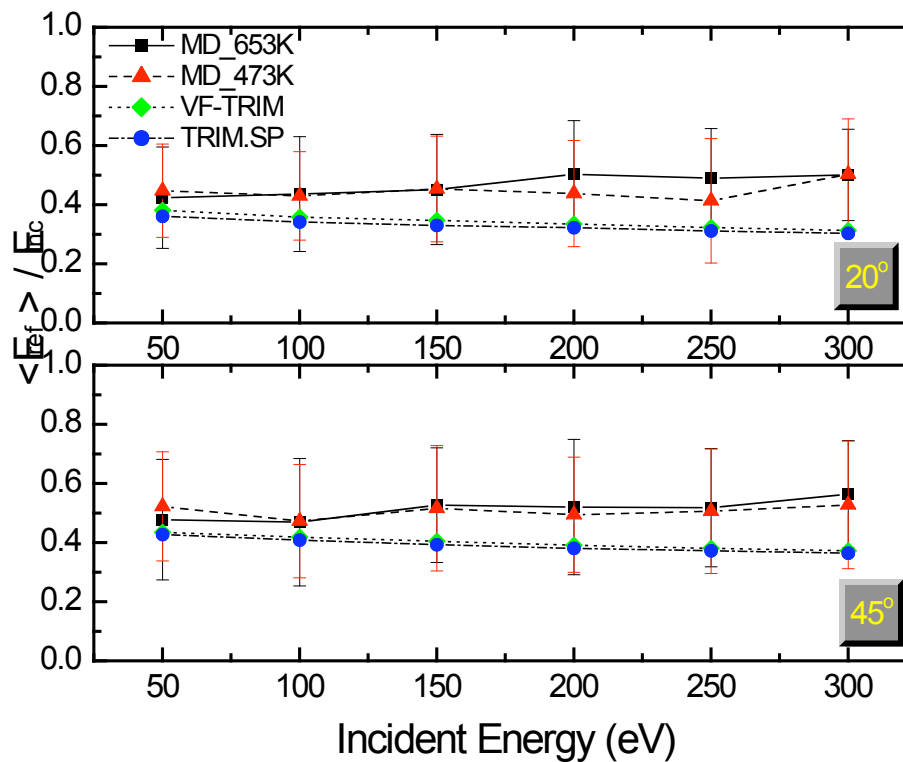
-- Reflection Coefficient



- Higher surface temperatures enhance reflection at moderate energies in MD
- Temperature effect is dominant at low energies and MD results with different temperatures converge at high energies
- Larger incident angles enhance reflection as expected
- MD and VF-TRIM & TRIM.SP show better agreement at higher energies
- The MD results at low energies need more flights for a better statistics

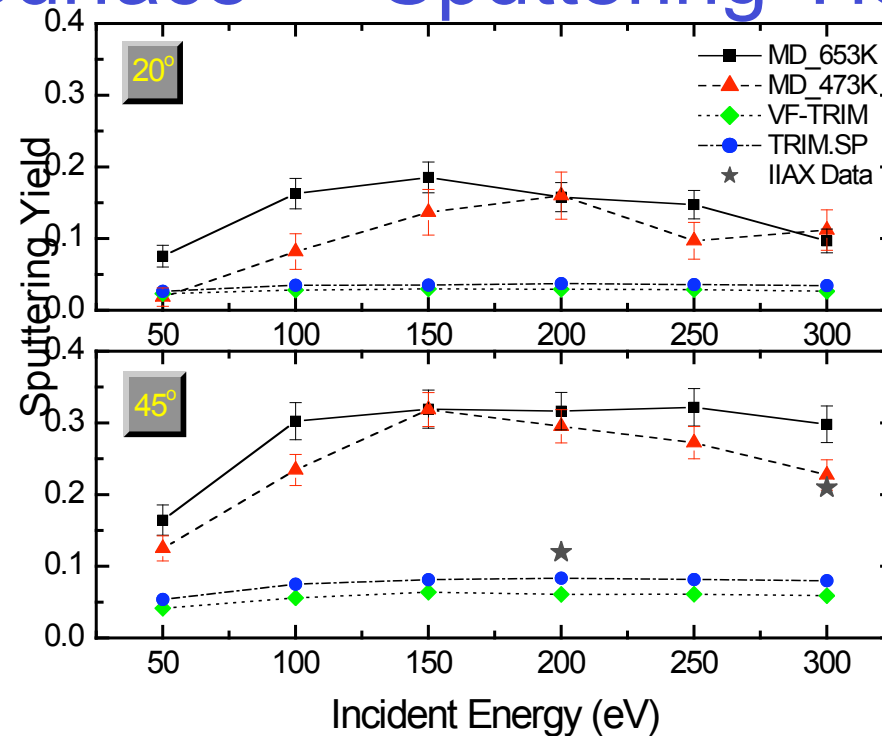
Reflection Results on Pure Li Surface

-- Average Reflected Energy



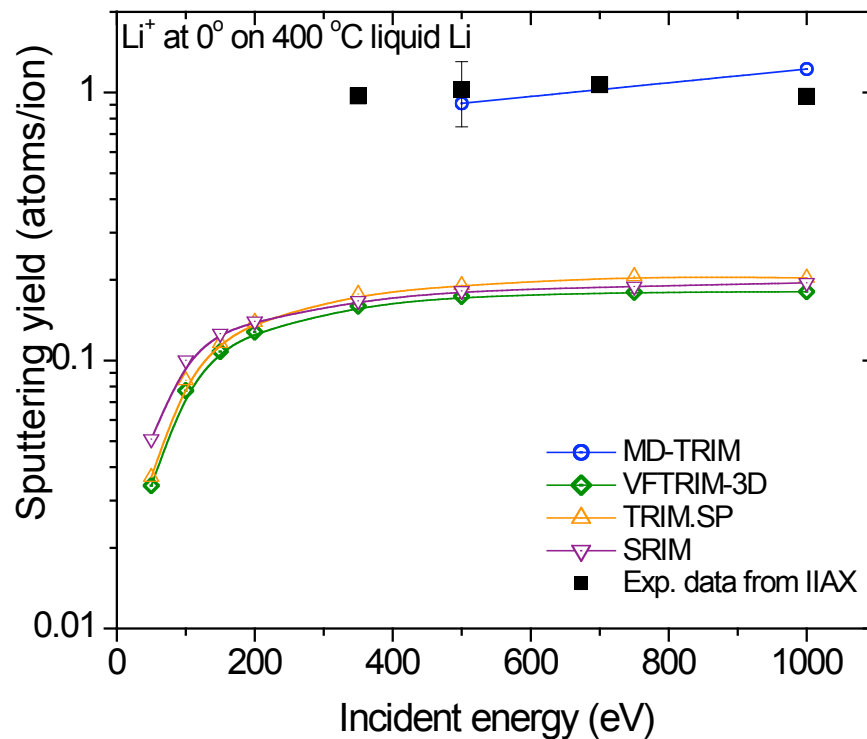
- Average energy of the reflected H atoms is independent of surface temperature
- Larger incident angles lead to larger average reflected energy as expected
- MD and VF-TRIM & TRIM.SP show a range of 30% ~ 50% for $\langle E_{ref} \rangle / E_{inc}$

Sputtering Results of D on Pure liquid Li Surface -- Sputtering Yield



- Higher surface temperatures enhance erosion in MD code
- Larger incident angles enhance sputtering in MD code
- Surface temperature has strong effect on the sputtering of liquid Li
- MD in liquid Li gets sputtered more compared with VF-TRIM & TRIM.SP in solid Li
- MD agrees with IIAX experimental data at high energy

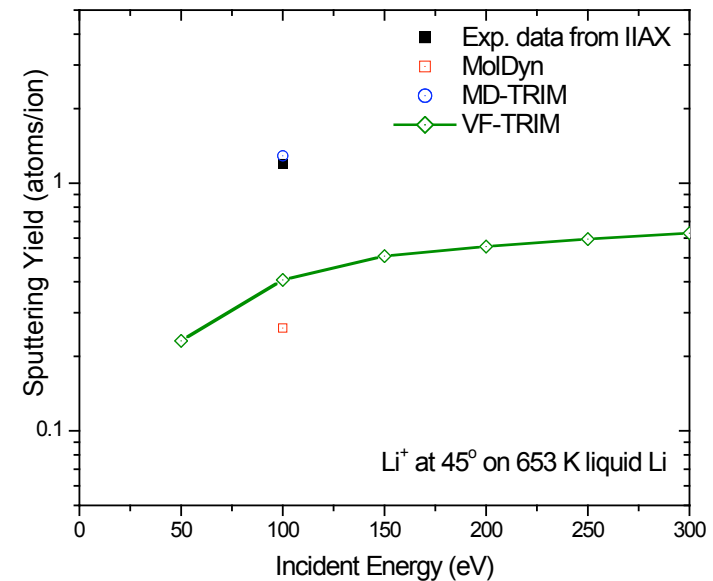
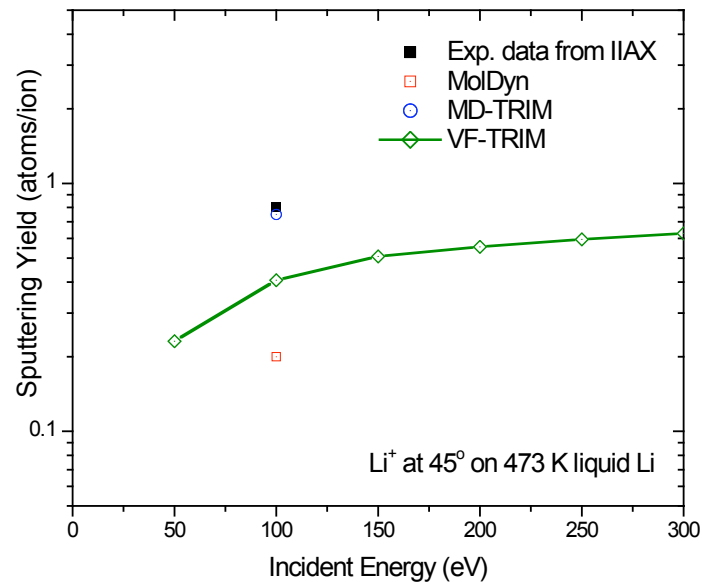
Need for MD-TRIM



Experimental and calculated sputtering yields for lithium self-sputtering of 400 °C liquid lithium at normal incidence.

- BCA codes
 - No concept of surface temperature
 - Cannot predict T dependent enhancement
- MD code
 - Needs a better surface potential
- MD-TRIM
 - VFTRIM calculation with
 - Recoil information from MD
 - SBE from MD
 - Matches experimental data

Comparison of models



Temperature (Kelvin)	Experiment (est.)	VF-TRIM	MolDyn	MD-TRIM
473	0.80	0.41	0.20	0.75
653	1.20	0.41	0.26	1.29

Surface Potential

- Current
 - Surface Binding Energy (SBE) consistency
 - Weighted surface potential acts $\pm 2.5\text{\AA}$ on the surface
 - CGP potential ^[6] for all Li atoms
- Future
 - Cohesive Energy consistency
 - LBP ^[9] (many-body) potential for bulk Li atoms
 - LJ ^[10] + LBP potentials for Li in surface layers
 - To make a smoother liquid Li surface
 - Stratification along the depth

^[9] Y. Li, E. Blaisten-Barojas and D. A. Papaconstantopoulos, *Phys. Rev.*, **B57** (1998-II) 519.

^[10] A. Rahman, *Physical Review*, **136** (1964) 405.

Future Modeling Work

- Develop a saturated LiH/LiD lattice and compare with the Li phase diagram
- Study H/D incident on saturated LiH/LiD surface and investigated the stability of the developed saturated LiH/LiD lattice
- Compare results b/w liquid pure Li and LiH/LiD surfaces
- Develop a better surface potential with consisting to the cohesive energy for the Li atoms within the surface layers
- Develop a better liquid Li surface with stratification structure along depth
- Investigate the effects of higher temperature on the sputtering and reflection which is leading to predict sputtering yield as a function of surface temperature
- Return to carbon and mixed material modeling

Conclusions

- IIAX – producing new data on Sn
- FLIRE – making first measurements of the retention of D from a plasma in a realistic divertor geometry
- Modeling – endeavoring to understand the experiments